



Carbon capture and biogas enhancement by carbon dioxide enrichment of anaerobic digesters treating sewage sludge or food waste



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HIGHLIGHTS

- The benefits of CO₂ enrichment on anaerobic digestion were evidenced.
- Sewage sludge and food waste anaerobic digesters were examined.
- First 24 h CH₄ production increased 11–16% for food waste and 96–138% for sludge.
- A mechanism of CO₂ utilisation has been hypothesised.
- Estimated potential CO₂ reductions of 8–34% for sludge and of 3–11% for food waste.

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ABSTRACT

The increasing concentration of carbon dioxide (CO₂) in the atmosphere and the stringent greenhouse gases (GHG) reduction targets, require the development of CO₂ sequestration technologies applicable for the waste and wastewater sector. This study addressed the reduction of CO₂ emissions and enhancement of biogas production associated with CO₂ enrichment of anaerobic digesters (ADs). The benefits of CO₂ enrichment were examined by injecting CO₂ at 0, 0.3, 0.6 and 0.9 M fractions into batch ADs treating food waste or sewage sludge. Daily specific methane (CH₄) production increased 11–16% for food waste and 96–138% for sewage sludge over the first 24 h. Potential CO₂ reductions of 8–34% for sewage sludge and 3–11% for food waste were estimated. The capacity of ADs to utilise additional CO₂ was demonstrated, which could provide a potential solution for onsite sequestration of CO₂ streams while enhancing renewable energy production.

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Abbreviations: AD, anaerobic digester; ANOVA, analysis of variance; CCS, carbon capture and storage; DC20, control digesters bubbled with N₂(g) for 20 min; DC0.5, control digesters bubbled with N₂(g) for 0.5 min; D0.3, digesters enriched with y_{CO₂} = 0.3; D0.6, digesters enriched with y_{CO₂} = 0.6; D0.9, digesters enriched with y_{CO₂} = 0.9; DI, deionized; DO, dissolved oxygen; GHG, greenhouse gases; H_i, Henry's constant for i; SAO, syntrophic acetate oxidation; sCOD, soluble chemical oxygen demand; TPAD, two phase anaerobic digestion; TS, total solids; UASB, upflow anaerobic sludge blanket; VFA, volatile fatty acid; VS, volatile solids; WWTP, wastewater treatment plant; k_{1L}, volumetric liquid phase mass transfer coefficient (s⁻¹); D_L, diffusion coefficient (m² s⁻¹); n, coefficient depending on the theory for interfacial mass transfer considered between the gas and the liquid phases; t₉₅, time to reach 95% of the equilibrium solubility (s); C*, solubility (mg L⁻¹); C₀, concentration at time zero (mg L⁻¹); C_t, concentration at time t (mg L⁻¹); (CO₂)_{generated}, CO₂ generated during the entire batch digestion process (mg); (CO₂)_{digestate}, CO₂ dissolved in the digestate at the end of the digestion period (mg); (CO₂)_{biogas}, CO₂ released with the biogas (mg); (CO₂)_{in}, CO₂ dissolved in the material to digest after the CO₂ injection (mg).

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1. Introduction

Carbon dioxide (CO₂) emissions to the atmosphere need to be reduced if targets for CO₂ reduction are to be met (e.g. UK [Climate Change Act, 2008](#)). Conventional carbon capture and storage (CCS) is based on the long term storage of this compound in geological or ocean reservoirs ([Xu et al., 2010](#)). This still has high associated costs and significant limitations linked to the potential risk of leaking from storage sites ([Holloway, 2007](#)). Moreover, the need to transport the CO₂ makes the proximity of source and reservoir a limiting factor. Therefore, the implementation of CCS is more feasible in large centralised sources which benefit from the pipeline's economy of scale ([Middleton and Eccles, 2013](#)).

The UK water industry emitted over 5 million tonnes of greenhouse gases (GHG) as CO₂ equivalents (CO₂e) during 2010–2011 ([Water UK, 2012](#)), of which 56% can be attributed to wastewater treatment ([DEFRA, 2008](#)). However, the varied size and scattered

location of organic waste and wastewater treatment plants (WWTPs), make the implementation of CCS particularly challenging in the water or waste sectors. This necessitates the development of alternative solutions for CO₂ capture and long term storage. Additionally, the increased implementation of upgrading technologies for the biogas produced in anaerobic digesters (ADs) (Weiland, 2010), results in the production of CO₂ concentrated streams. This further raises the need to develop new carbon storage or utilisation technologies applicable to the wastewater and waste sectors.

Biogenic carbon sequestration methods (e.g., microalgae, biochar) are being studied as alternatives to geological or oceanic reservoirs. However, in general, their capacity for CO₂ sequestration or their large-scale applicability needs to be further investigated (NERC, 2011). A few studies have considered the potential of CO₂ biological conversion in anaerobic processes, reporting benefits both in terms of carbon uptake and renewable energy production. Alimahmoodi and Mulligan (2008) stated a 69–86% CO₂ uptake when dissolving this gas in the influent to an upflow anaerobic sludge blanket (UASB) reactor. Salomoni et al. (2011) further confirmed the potential of CO₂ biological conversion in two phase anaerobic digestion (TPAD), and observed a 25% methane (CH₄) yield enhancement when bubbling CO₂ into the first stage. Sato and Ochi (1994) stated associated benefits of up to 30% increased specific CH₄ yields when enriching ADs treating sewage sludge with CO₂.

Therefore, the capacity of ADs to transform CO₂ into CH₄ could result in the onsite treatment of CO₂ concentrated streams and potential increases in CH₄ production. Although the benefits of CO₂ enrichment of ADs have been evidenced, the scarcity of the literature available requires further research before its full potential can be estimated. Furthermore, the increasing practice to treat food waste or mixed substrates, also needs to be considered in relation to the benefits of CO₂ enrichment.

This paper assessed the impact of CO₂ injection in batch ADs treating food waste or sewage sludge. Renewable energy production, CO₂ utilisation and digestate quality were studied. Firstly, absorption tests were completed to estimate the gas–liquid contact time required to reach CO₂ equilibrium conditions between the liquid phase and the injected gas. Secondly, the impact of CO₂ enrichment in batch ADs treating food waste and sewage sludge was assessed for CO₂ molar fractions (y_{CO_2}) of 0.3, 0.6 and 0.9 (0.4, 0.8 and 1.6 bar CO₂ partial pressures [p_{CO_2}]). Lastly, the time required to recover from any initial acidification due to CO₂ injection was determined for sewage sludge by monitoring the pH of sacrificial ADs.

2. Methods

2.1. Description of the anaerobic digester equipment

Each batch AD unit consisted of a 1 L glass bottle with a four port cap (Fisher Scientific, Loughborough, UK). Two ports were used for gas injection by means of Pyrex diffusers with a porosity of 3 and 15 mm diameter (Fisher Scientific, Loughborough, UK). When running absorption tests, one port was acting as pressure release and the fourth port was blocked (Fig. 1a). When conducting CO₂ enrichment tests in ADs, one port was blocked with a 17 mm septa (Thames Restek UK Ltd., Buckinghamshire, UK), allowing gas sample extraction for composition analysis, and the last port was connected to a MilliGascounter (Litre Meter Ltd., Buckinghamshire, UK) for biogas volume recording (Fig. 1b). When running sacrificial ADs for pH monitoring, one port was used for daily sample extraction of the liquid phase. The ADs were continuously stirred and placed in a temperature controlled water bath (38 ± 0.5 °C).

2.2. Absorption tests methodology

The contact time required to ensure CO₂ equilibrium conditions between the gas injected and the sewage sludge or food waste, was estimated by conducting oxygen (O₂) absorption tests with air, and converting the results to CO₂ using diffusion coefficients, as previously suggested by Garcia-Ochoa and Gomez (2009). In order to account for the viscosity variability of food waste and sewage sludge, tests with different liquid viscosities were performed. Glycerol was used as a viscosity enhancer, because of the extensive information available of its impact on aqueous solutions (Jordan et al., 1956). Tests in deionized (DI) water with air flow rates of 0.5, 1.0 and 1.7 L min⁻¹ and tests with a fixed air flow rate (1.0 L min⁻¹) and mixtures of glycerol in DI water of 10%, 30%, 50% and 70% weight (glycerol ≥ 98%; Fisher Scientific, Loughborough, UK) were performed. The air flow rate was controlled by a mass flow controller (MFC) (Premier Control Technologies, Norfolk, UK). The dissolved oxygen (DO) was monitored using a DO probe (HACH LDO101; Camlab, Cambridge, UK) connected to a meter device (HACH HQ30d; Camlab, Cambridge, UK).

The gas to liquid mass transfer was described using Eq. (1) and corrected for the time to reach 95% of the equilibrium solubility by Eq. (2). Considering similar equations for CO₂ and O₂, and relating the volumetric mass transfer coefficients ($k_L a$) of both gases with the ratio of their diffusion coefficients (Eq. (3)), a relationship between the times to reach equilibrium solubility with CO₂ and with O₂ was obtained (Eq. (4)). The film theory for interfacial mass transfer was considered, which states $n = 1$. The diffusion coefficients for CO₂ in water-glycerol mixtures used in Eq. (4) were 2.6 × 10⁻⁵, 1.7 × 10⁻⁵, 7.2 × 10⁻⁶ cm² s⁻¹ for glycerol concentrations of 0%, 25% and 50% weight, respectively. The values used for O₂ were 3.0 × 10⁻⁵, 3.4 × 10⁻⁵, 1.6 × 10⁻⁵ cm² s⁻¹ for glycerol concentrations of 0%, 25% and 50% weight, respectively. These diffusion coefficients were obtained from those reported by Brignole and Echarte (1981) and Jordan et al. (1956), for CO₂ and O₂, respectively, after correction for a temperature of 38 °C as per Diaz et al. (1987).

$$\ln \left(\frac{C^* - C_t}{C^* - C_0} \right) = -k_L a \cdot t \quad (1)$$

$$\ln(0.05) = -k_L a \cdot t_{95} \quad (2)$$

$$(k_L a)_{CO_2} = (k_L a)_{O_2} \cdot \left[\frac{(D_L)_{CO_2}}{(D_L)_{O_2}} \right]^n \quad (3)$$

$$(t_{95})_{CO_2} = (t_{95})_{O_2} \cdot \frac{(D_L)_{O_2}}{(D_L)_{CO_2}} \quad (4)$$

where $k_L a$: volumetric liquid phase mass transfer coefficient (s⁻¹), D_L : diffusion coefficient (m² s⁻¹), n : coefficient depending on the theory for interfacial mass transfer considered between the gas and the liquid phases, t_{95} : time to reach 95% of the equilibrium solubility (s), C^* : solubility (mg L⁻¹), C_0 : concentration at time zero (mg L⁻¹), C_t : concentration at time t (mg L⁻¹).

2.3. Methodology for enriching the digesters with CO₂

Batch ADs treating food waste or sewage sludge were operated with an inoculum to substrate volatile solids (VS) ratio of 2:1 and a working volume of 700 ml. Macerated and digested food waste were collected from a full scale UK AD site treating 30,000 tonnes of organic waste per year. Thickened waste activated sludge (WAS) and digested sewage sludge were collected from a full scale UK WWTP serving a 2.5 million population equivalent.

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